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PERSISTENCE OF PLUTONIUM IN SOIL, PLANTS AND SMALL MAMMALS*

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Abstract--A 10-year surveillance was made of the persistence of residual ^{239}Pu in soil, plants and small mammals indigenous to fallout areas contaminated with ^{239}Pu dispersed by high explosive detonations. Downward migration of fallout particles occurred in undisturbed soil profiles, and wind and water erosion accounted for some redistribution of the initial ^{239}Pu contamination. Long-term cropping experiments showed a relatively low degree of ^{239}Pu transfer from soil to plants but a consistent increase in its accumulation in plant tissue during a 5-year cropping sequence. Synthetic chelating agents enhanced plant uptake of ^{239}Pu from soil. The accumulation of residual ^{239}Pu in kangaroo rats and jackrabbits was highest in bone tissue; considerable amounts also were found in lung tissue. High levels found in the gastrointestinal tract indicate that ingestion is a major pathway through which residual ^{239}Pu entered into these small mammals.

INTRODUCTION

Several high explosive detonations involving ^{239}Pu were set off at the Nevada Test Site (1955-1957) to evaluate various problems that might arise from an accidental or experimental detonation of atomic weapons, while little or no fission occurs. In analyzing the health hazard arising from the ^{239}Pu dispersed by such detonations, Langham and coworkers [(1)] concluded that the fallout conditions create an immediate or acute inhalation hazard to anyone caught in or entering the fallout area before all of the particulate matter has settled to the ground. The magnitude of this acute hazard depends upon the concentration of airborne ^{239}Pu, the respiratory rate of the individual, the time of exposure, and to some extent the particle size distribution of the radioactive fallout material. Data on the airborne concentration of ^{239}Pu following detonation have been reported by Shreve[(2)], Wilson, et al.[(3)], Olafson and Larson [(4)], and Mork [(5)].

Langham and coworkers further concluded that the residual surface contamination creates a potential ^{239}Pu inhalation hazard to persons living or working in the fallout area for a relatively long time after the detonation; however, they emphasized strongly that the deposited ^{239}Pu constitutes no hazard whatsoever so long as it remains deposited. This residual hazard, therefore, depends upon that fraction of ^{239}Pu which is subsequently disturbed and resuspended in such a manner as to gain entrance into the body. Some findings on the mechanical disturbance and resuspension of ^{239}Pu deposited in the fallout area were reported by Mork [(5)]. It is possible, but certainly not economical, to virtually eliminate this residual hazard by removing contaminated

crops, replacing topsoil, and deep plowing farmland as was done after an aircraft accident near Palomares, Spain, in January 1966, which resulted in some ^{239}Pu contamination.

Inasmuch as contamination and ingestion of food may contribute to the residual ^{239}Pu hazard, a surveillance of ^{239}Pu transfer from soil to biotic components in the fallout area should provide information for evaluating its significance with passing time. The findings reported herein indicate the persistence of residual ^{239}Pu in soil, plants, and small mammals indigenous to fallout areas during the decade following its deposition.

MATERIALS AND METHODS

The geographic location of the two fallout areas involved in this study was described by Paglia [(6)]. Casual field surveys were conducted in October 1956, July 1958, and June 1966. Details of the methods for collecting and processing samples have been reported [(7)]. Briefly, kangaroo rats (_Dipodomys microps_ and _D. merriami_) and jack-rabbits (_Lepus californicus_) were separated into bone, muscle, gastro-intestinal tract and contents, and lung fractions before ashing them for ^{239}Pu analysis. Plant samples were dried, ground, and ashed in preparation for analysis. Soil samples were collected for ^{239}Pu analysis and also for further experimental work under laboratory conditions. A 5-year cropping experiment with ladino clover (_Trifolium repens_) began in 1958; tests for the effects of chelating agents on ^{239}Pu uptake by alfalfa (_Medicago sativa_) were conducted in 1966-1968. The ^{239}Pu concentration in samples was determined by the TTA extraction

procedure of Eisenacher adapted for soil, plants, and animal tissues [8].

RESULTS AND DISCUSSION

Considerable heterogeneity occurred in the particle size distribution of fallout and in the subsequent levels of residual ^{239}Pu deposited at sampling stations downwind from the high explosive detonations. This residual contamination became stabilized within a few weeks after fallout deposition occurred and the resuspension of particulate matter containing ^{239}Pu was practically nil, except from heavy gusts of wind and mechanical disturbance. The rapid attenuation of the alpha activity initially measured on the soil surface indicated downward migration of fallout particles in contrast to observations at the Trinity fallout area, in New Mexico, where surface alpha activity levels remained unchanged with little or no evidence of downward migration during one decade of time following depositions [8]. Differences in the types of detonation and the kinds of fallout particle deposited at the soil surface are believed to account for this variation.

X-ray diffraction patterns showed that the clay materials of soil in the Nevada fallout patterns are predominantly montmorillonitic and illitic. Alternate freezing and thawing, and alternate wetting and drying, cause a granulating action in this type of soil which involves the processes of aggregation and dispersion. These processes, in turn, are conducive to mechanical movement of high density particles. Downward migration of ^{239}Pu-contaminated fallout particles also could occur with the flow of infiltrating water. Christensen and Thomas [9]

(Table 1) reported the penetration of Pu-species to at least 28 feet along fissures in Los Alamos tuff. The data in Table 1 are typical of the distribution of ^{239}Pu in profiles of undisturbed soil where downward migration to depths varying from 6 to 9 cm had occurred during the decade after it was deposited. The effectiveness of wind and rainfall erosion on redistribution of fallout particles was commensurate with the intensity of these natural forces in the fallout areas during the period of this study; high concentrations of alpha activity found in wind-blown material and in stream sediments indicated considerable fallout particle movement by wind and water erosion. Because of the high degree of adsorption of Pu-polymers known to occur in soil [10, 11], and its slow rate of diffusion and leaching by invading ground water [12], it would appear unlikely that residual ^{239}Pu could move downward in soil in solution form.

An assessment of soil to plant transfer of ^{239}Pu through roots could not be made in the field because of the possible contamination of foliage from wind-blown material. Earlier studies [13] showed that fallout particles are difficult to remove from contaminated leaf surfaces. Accordingly, long-term cropping experiments were conducted under glasshouse conditions in which ladino clover was grown on soil containing ^{239}Pu-contaminated fallout debris. The results of plant

(Table 2) uptake through roots during a 5-year study are shown in Table 2. The total crop yields increased each year with a subsequent increase in the total amount of ^{239}Pu removed by plants from the soil. The accumulation of ^{239}Pu in plant tissue increased from 3 d/m/g to about 23 d/m/g during the 5-year cropping sequence. Some of this increase

may be attributed to the continuing development of the root system until the crop became fully established, forcing more intimate contact of roots with ^{239}Pu-contaminated fallout particles. The possibility cannot be overlooked, however, that residual ^{239}Pu may have become more available for root uptake with passing time. Newbould [14] reported slightly higher ^{239}Pu uptake in the second year crop than in the first year crop of rye grass grown on three contaminated soils. Another factor for consideration is that chelating materials are known to enhance plant uptake of heavy metals [15] and that some of these kinds of organic compounds may naturally occur in soil. Table 3 shows the results of an experiment to test the effects of two synthetic chelating agents on ^{239}Pu uptake by alfalfa. The DTPA chelate significantly increased plant uptake of residual ^{239}Pu, whereas EDDHA chelate was less effective at levels of 100 ppm in soil on the dry weight basis. Our findings agree with those of other investigators[16, 17, 18] which indicate that relatively small amounts of ^{239}Pu are transferred from soil to plants through roots. Samples of native plants collected from fallout areas contained higher levels of ^{239}Pu, virtually all of which we must assume was from external surface contamination.

(Table 3)

Collections of small mammals were not always good during the casual field surveys. We took what could be found at or near the sampling stations in the fallout areas during one to three day collection periods. Table 4 shows some findings on the persistence of ^{239}Pu in tissues of kangaroo rats and jackrabbits collected in 1956, 1958, and 1966. The data for kangaroo rats are from pooled samples collected

(Table 4)

in a trapping grid. This was necessary to obtain samples of sufficient size for ^{239}Pu analysis, but it did not permit an evaluation of the variation in tissue burden for single animals. There was, however, considerable variation in tissue burdens of individual jackrabbits. For example, five mature animals collected near sampling station 13-3 contained 0.27, 0.28, 0.57, 2.11 and 0.03 disintegrations per minute of ^{239}Pu per gram of bone tissue. Their gastrointestinal tract contents showed greater variation, reflecting that which had been ingested during the three day period prior to collection. Muscle tissue was analyzed, but only trace amounts could be detected in any of the samples. No gross anatomic lesions were detected in these small mammals. Paglia [(6)] reported results of hematopathological surveys made on the kangaroo rats from these fallout areas.

The accumulation of residual ^{239}Pu was highest in bone tissue; considerable amounts also were found in lung tissue, especially from animals living in areas of high residual ^{239}Pu contamination. The high levels found in the gastrointestinal tract indicate that ingestion is a major pathway through which the residual ^{239}Pu entered into these small mammals. The most probable source of this contamination was fallout particles swallowed when eating or while preening fur after digging in soil and taking dust baths. The deposition of ^{239}Pu in bone tissue not only continued to persist with passing time but it also appeared to have increased relative to the lung tissue burden and ingestion level at the time of sacrifice. This raises the question of whether or not residual ^{239}Pu becomes more biologically available through weathering processes with passing time; however, more detailed

and intensive survey work in the fallout area is needed to determine the extent to which this might occur. We have found no literature references indicating an increased solubility of ^{239}Pu-fallout particles upon weathering with passing time. The laboratory experiments of Katz, et al.[19], in which Pu was administered orally to rats in chronic, low-level feedings, indicated that less than .003 per cent of the ingested dose was absorbed from the gut of which about 90 per cent was deposited in the bone. In projecting conditions affecting human health hazards, Langham, et al.[1] suggest that from 2 to 10 per cent of the inhaled dose could be absorbed from the lung, with about 80 per cent of this subsequently being fixed in the skeleton. The effective half time of ^{239}Pu in man was estimated by Langham to be about 200 years[20]. In a study of cattle that had been grazing on or near the Project Rollercoaster sites where ^{239}Pu had been liberated, Brechbill[21] found the highest levels of ^{239}Pu in bone tissue and significant levels in spleen and lung tissues. These levels, however, were far below the maximum permissible concentration levels established for humans. The results indicate that ingestion is the major route through which ^{239}Pu entered cattle grazing at these contaminated sites.

Because of limited access into the fallout areas and limited availability of animals, our casual field surveys did not involve an intensive study of residual ^{239}Pu uptake by a given population of small mammals living in a defined area. Consequently, these findings represent only qualitative trends in the persistence of residual ^{239}Pu in bone and lung tissues with passing time. The

living habits of these small mammals are such that they are in much more intimate contact with the residual ^{239}Pu than would be the case for humans living in such a contaminated fallout area; therefore, these findings probably represent extremes in the transfer of residual ^{239}Pu contamination <u>via</u> inhalation and ingestion.

Table 1. Distribution of residual ^{239}Pu in undisturbed soil profiles

Profile depth cm.	Profile 1 d/m/g	Profile 2 d/m/g	Profile 3 d/m/g
0-3	2,860	12,050	6,200
3-6	163	2,440	1,710
6-9	21	344	545
9-12	54	353	215

Table 2. ^{239}Pu in successive crops of ladino clover grown on contaminated soil under glasshouse conditions

Cropping period	Crop yield g. dry wt.	^{239}Pu in dry plant material d/m/crop	d/m/g
1958	66 (3)[a]	205	3.1
1959	110 (4)	737	6.7
1960	194 (5)	1377	7.1
1961	310 (6)	3565	11.5
1962	316 (6)	7142	22.6

[a] Number of cuttings harvested at the quarter-bloom stage each year. The potting containers were 60-liter steel drums containing 1:1 mixtures of Yolo soil and Area 11 soil (.05 m^3 soil volume; .12 m^2 surface area). The residual ^{239}Pu level was 1.18 ug/g soil (1.62×10^5 d/m/g).

Table 3. Influence of chelating agents on ^{239}Pu uptake by alfalfa grown on contaminated soil

Soil	Control	DTPA	EDDHA
		d/m/gram dry plant*	
11-N	0.67	2.77	0.35
11-C	0.59	7.80	0.41
11-D1	0.34	7.25	1.93
11-D2	3.25	12.10	5.75

* Mean from 3 replicates of potted soils (9 kg. lots); DTPA (diethylenetriamine pentaacetic acid) and EDDHA (ethylenediamine di-o-hydroxyphenylacetic acid) were mixed with surface soil (3 cm layer) at 100 ppm dry weight basis. The residual ^{239}Pu level in these soils was not determined.

Table 4. Mean levels of ^{239}Pu in tissues of small mammals indigenous to fallout areas

Survey station	Residual ^{239}Pu	Bone	GI. tract	Lung
	d/m/g. soil		d/m/animal part	
1956	KANGAROO RAT (Dipodomys microps and D. merriami)			
S-61 (6.4)[a]	42 (1)[b]	.60 (64)[c]	16	.02
S-88 (16)	200 (1)	.38 (30)	67	.11
1958				
S-61 (6.4)	22 (2.8)	.65 (6)	84	.07
11-D (0.2)	12,054 (2.8)	7.13 (10)	2,052	11.40
13-2 (0.8)	241 (1.3)	4.30 (20)	1,255	.12
13-3 (4.8)	20 (1.3)	.20 (5)	252	.10
13-5 (6.4)	5 (1.3)	.14 (16)	108	.07
1966				
11-D (0.2)	6,200 (10.8)	47.05 (11)	1,050	61.28
13-3 (4.8)	4 (9.3)	2.72 (38)	170	5.80
1958	JACKRABBIT (Lepus californicus)			
13-1 (0.2)[a]	2,860 (1.3)[b]	128.48 (6)[c]	5.5×10^5	57.50
13-2 (0.8)	241 (1.3)	11.68 (7)	3.2×10^4	.36
13-3 (4.8)	20 (1.3)	1.75 (5)	5,712	.24
13-5 (6.4)	5 (1.3)	.58 (8)	2,490	.11
1966				
11-D_1 (0.2)	6,200 (10.8)	665.40 (2)	4.1×10^4	98.25
11-D_2 (1.4)	673 (10.8)	88.76 (2)	1.6×10^4	8.92
13-3 (4.8)	4 (9.3)	19.27 (5)	1,360	1.92
13-5 (6.4)	- (9.3)	2.34 (2)	781	.10

[a] Distance (km.) downwind from ground zero.

[b] Duration (year) of residual ^{239}Pu contamination.

[c] Number of animals from which mean was derived.

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